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Soft water-permeable polyolefins nonwovens having opaque characteristics.

A method for making nonwoven material containing polyolefin filaments and having an increased degree of opacity, characterized in that at least about 25% of the total weight of the filaments in the nonwoven web are polyolefin filaments that have a delta "\Delta" or diamond cross-sectional configuration, an initial spun denier not exceeding about 24 dpf, and a final drawn denier of not less than about 1 dpf is disclosed.

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## SOFT WATER-PERMEABLE POLYOLEFINS NONWOVENS HAVING OPAQUE CHARACTERISTICS

This invention relates to a method for nonwoven material containing polyolefin filaments having particular cross-sectional configurations.

The chemical inertness, low allergenic properties, high tensile strength and low melting point of polyolefin fiber and filaments makes them useful in nonwoven materials from which personal contact products such as cover-stock for diapers can be made, as described, for instance, in U.S. Patents 4,112,153, 4,391,869, 4,573,987, and 4,578,066.

Such materials must be cost-competitive, retain substantial cross directional (CD) strength and toughness, and have a soft surface "feel". However, an effective combination of such properties is difficult to achieve in nonwovens with existing technology and using conventional synthetic fibers, as described in the said patents. In particular, softness is usually gained at the expense of cross directional strength and a substantial increase in cost.

In the case of personal-contact products, such as diaper cover-stock, and other covering purposes, it is also desirable to enhance certain non-functional esthetic properties, such as opacity and stain-masking ability. In order to achieve such properties, including a preferred degree of opacity of 32%-45%, it is even more difficult to provide an acceptable balance of properties, particularly with chemically inert polyolefins such as polypropylene.

Colorants and brighteners have been used as spun melt components to enhance opacity and stainmasking ability, but they cause additional problems such as leaching, allergenic reactions and increased cost.

A method for making nonwoven material containing polyolefin filaments that increases its opacity while reducing the need for high concentrations of colorants is therefore needed.

According to the invention, a method for making nonwoven material containing polyolefin filaments, comprising assembling a web of filaments, including the polyolefin filaments, and bonding the filaments to form a nonwoven material, is characterized in that at least about 25% of the total weight of the filaments in the web are polyolefin filaments that have a delta " $\Delta$ " or diamond cross-sectional configuration, an initial spun denier not exceeding about 24 dpf, and a final drawn denier of not less than about 1 dpf.

Generally, by the method of the instant invention, one can achieve an opacity within the range of 32%-45% or even higher, depending upon the balance of interdependant properties selected for the final product.

It is possible to obtain nonwoven materials in a wide range of weights, from heavy to as light as 10-30 gm/yd² and having substantially improved opacity and stain-hiding properties without substantial sacrifice in other areas.

The production techniques for obtaining the various polyolefin cross-sectional configurations, and the conventional methods for producing the nonwovens material itself, are well known in the art and are not part of the present invention. Thus, conventional techniques for bonding the filaments to form nonwoven material, such as spun bonding, needle punching and thermal or sonic bonding techniques may be used. However thermal bonding is generally the most effective fabrication technique to obtain a wide range of weights at low cost.

In the method according to the invention, the web of filaments can comprise, in addition to the polyolefin filaments of delta or diamond cross sections, other types of conventionally used filaments such as other polyolefin filaments or rayon filaments, including filaments having various known cross-sectional configurations, such as "y", "x", "o" (round), oval, square, and rectangular configurations, including blends of such filaments with fibrillated film (such as polyolefin film). The particular combination and amount of filament of delta or diamond configuration within the limits required according to the invention, will depend substantially upon the degree of opacity desired in combination with other properties, such as strength and a soft or velvety feel.

Preferably, the proportion of delta and/or diamond cross-sectional configuration to another cross-sectional configuration is about 50%, with the other 50% configuration preferably being round.

Also preferably, the delta cross-sectional polyolefin filament have a preferred initial sum denier within a range of about 2.0-4.0 dpf and a final drawn denier corresponingly within the range of about 1.0-3.0 dpf, more preferably 1.9-2.5 dpf, in order to retain both strength and softness.

Preferably, for a combination of softness and cross-dimensional strength, polyolefin filaments having both delta and diamond cross-sectional configurations are used. The desired combination may be supplied by a uniform blend in a single web or laminated group of webs of uniform composition, or by a plurality of homogeneous webs individually differing in the blend of filaments used.

Preferably, the filaments used vary from about 2.5-7.6 cm in length. Longer filaments naturally produce higher cross directional tensile strength, and mixtures of long and short filaments within the above range tend to provide optimum toughness. For example, a 50:50 mixture of 2.5 cm diamond with longer (e.g. 3.8cm-5cm) round cross-sectional filament is preferred for providing both strength and a velvet-like feel.

The following examples and table further illustrate the present invention.

## Example 1

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- A. Delta cross-sectional isotactic polypropylene filament of 4.0 dpf spun denier is produced in a conventional manner by melt spinning at 290°C using PRO-FAXR 6501 polypropylene polymer (commercially available from Hercules Incorporated of Wilmington, Delaware), degraded in the usual way with .025% Lupersol to an MFR (Melt Flow Rate) (ASTM D 1238-82) value of 16 and spun, using a 700 hole delta spinnerette to obtain a final drawn denier of 2.1 dpf. Crimped (10 crimps/cm) bundles are then cut into 2.54 cm length, collected, and compressed into bales for later testing.
- B. Round cross-sectional polypropylene filament of 2.8 dpf spun denier is similarly produced in a conventional manner by melt spinning PRO-FAX<sup>R</sup> 6501 polypropylene polymer degraded to an MFR value of 13, spun at 290 °C to obtain a final drawn denier of 2.1 dpf, crimped as before, cut into 2 inch lengths, collected, compressed and baled for later testing.
- C. Delta cross-sectional polypropylene of 2.6 dpf spun denier is produced by melt spinning at 285°C, using PRO-FAX 6301 (commercially available from Hercules Incorporated of Wilmington, Delaware), and finally drawn to 2.2 dpf. comped as before, cut into two inch (2") bundles, collected, compressed, and baled for later testing.
- D. Delta cross-sectional fiber of Example 1 A (2.1 dpf denier) is crimped as before and cut into 1.5 inch bundles collected and compressed into bales for later testing.
- E. Round cross-sectional fiber of 2.8 dpf spun denier is drawn to 2.1 dpf as in Example I B, crimped as before and cut into 3.8 cm bundles, collected, and compressed into bales for later testing.
- F. Staple cut fiber of delta and round cross-sectional configuration treated as described in C. and B. supra is combined in a homogeneous ratio of 50-to-50 parts by weight, collected, compressed and baled for later testing.
- G. Round cross-sectional polypropylene filament of 1.5 dpf is produced in the manner of Example 1 B by melt spinning PRO-FAX 6501 polypropylene polymer degraded to an MFR value of 12 at 285 °C and drawn to obtain a final drawn denier of 1 dpf, crimped as before, cut into 1.5 inch lengths, collected, compressed and baled for later testing.
- H. Delta cross-sectional polypropylene of 1.5 dpf spun denier is produced the manner of Example I C by melt spinning PRO-FAX 6501 at 285 °C and drawn to 1.0 dpf, crimped as before, cut into 3.8 cm bundles, compressed, and baled for later testing.
- I. Round cross-sectional polypropylene filament of 8.0 dpf is produced from the same melt and in the manner of Example I B, spun to obtain a 6 dpf final denier, crimped crimped as before, cut into 3.8 cm, lengths, collected, compressed, and baled for later testing.

## Example 2

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- A. Bales 2.5 cm crimped polypropylene staple of delta cross-sectional configuration as described in Example I A is broken, and formed into two identical homogeneous webs in a conventional manner, and the webs superimposed in machine direction as they are transferred onto a continuous fiber glass belt, and thermally bonded, using a hot diamond-patterned calendar at 165° C and 276 kPa (40 psi) roll pressure to a obtain a nonwoven weighing 20gm/yd². The resulting material, identified as NW-1, is then cut into convenient dimensions for conventional testing purposes and test results reported in Table I below (in which Table and similar tables the values corresponding to 20 gm/m² are obtained by multiplying by 0.9 the values corresponding to 20gm/yd²).
- B. Baled 5 cm crimped polypropylene staple of round cross-sectional configuration as described in Example I B is broken, and formed into two identical homogeneous webs in a conventional manner, the webs being superimposed in machine direction as they are transferred onto a continuous fiber glass belt, and thermally bonded as in Example 2 A, using a hot diamond-patterned calendar to obtain a semi-opaque non-woven weighing 20gm/yd². The resulting material, identified as NW-2, is then cut into convenient

dimensions for testing purposes, standard tests run, and test results reported as control in Table I below.

- C. The 2.5 cm and 5 cm crimped staple of delta and round configuration of Examples I A and I B is added to separate openers and conveyed into separate cards to form two homogeneous webs with a 25/75 weight ratio of 1" delta/2" round in a conventional manner, the webs being transferred onto a continuous fiber glass belt, and thermally bonded as before, using a hot diamond-patterned calendar to obtain a nonwoven material weighing 20.7gm/yd². The resulting material, identified as NW-3, is then cut into convenient dimensions for testing purposes, standard tests run, and test results reported in Table I below.
- D. The 2.5 cm and 5 cm crimped staple of Examples I A and I B is added to separate openers, broken, conveyed into separate cards, and formed into two homogeneous webs having a 50/50 ratio of 2.5 cm delta/5 cm round in a conventional manner, the webs being superimposed in machine direction as they are transferred onto a continuous fiber glass belt, and thermally bonded as before, using a hot diamond-patterned calendar to obtain a nonwoven material weighing 20.7gm/yd². The resulting material, identified as NW-4, is then cut into convenient dimensions for testing purposes, standard tests run, and test results reported in Table I below.
- E. The 2.5 cm and 5 cm crimped staple of Examples I A and I B is added to separate openers, broken and conveyed into separate cards and formed into two identical homogeneous webs of 2.5 cm delta/5 cm round of 75/25 weight ratio in a conventional manner, the two webs being superimposed in machine direction, transferred onto a continuous fiber glass belt, and thermally bonded as before, using a hot diamond-patterned calendar to obtain a nonwoven material weighing 19.3gm/yd². The resulting material, identified as NW-5, is then cut into convenient dimensions for testing purposes, standard tests run, and test results reported in Table I below.
- F. Baled combined 5 cm crimped staple of 50:50 delta:round cross-sectional configuration by weight, as described in Example I F (1 B and 1 C) is broken and formed into two identical mixed fiber webs in the same general manner as before, the webs being superimposed in machine direction, transferred onto a continuous fiber glass belt, and thermally bonded as before, using a hot diamond-patterned calendar to obtain a nonwoven material weighing 19.1gm/yd². The resulting material identified as NW-6 is then cut into convenient dimensions for testing purposes, standard tests run, and test results reported in Table I below.
- G. Baled 3.8 cm crimped staple of drawn 2.1 dpf delta cross-section, as described Example I D is broken and formed into a web in the same manner as before. A second web is then prepared using 3.8 cm crimped staple of 2.1 dpf circular cross-section as described in Example IE is broken and formed into a web of equal weight in the same manner as before.
- The two webs, consisting of different fiber cross-section are superimposed in a machine direction, transferred onto a continuous fiber glass belt, and thermally bonded as before, using a hot diamond-patterned calendar to obtain a nonwoven material weighing 18gm/yd². The resulting material identified as NW-7 is then cut into convenient dimensions for testing purposes, standard tests run, and test results reported in Table I below.
- H. Baled 3.8 cm (1.5") polypropylene staple of round cross-sectional configuration (extruded 1.5 dpf drawn 1 dpf) as described in Example 1 G is broken and formed into two identical homogeneous webs, the webs being superimposed in machine direction as they are transferred onto a continuous fiber glass belt then thermally bonded, using a hot diamond-patterned calendar at 165°C and 276kPa (40 psi) roll pressure to obtain a nonwoven weighing 20gm/yd². The resulting nonwoven, identified as NW-8, is then cut into convenient dimensions for testing purposes, and test results reported in Table I below as a control.
- I. Baled 3.8 cm polypropylene staple of delta cross-sectional configuration drawn to 2.1 dpf from Example 1D, and round cross sectional configuration from 1E, are combined in the manner of Example 2 G supra to obtain an opaque nonwoven weighing about 20gm/yd². The resulting material, identified as NW-9, is then cut into convenient dimensions for testing purposes and test results reported in Table I below as a control.
- J. Baled 3.8 cm polypropylene staple of round cross-sectional configuration and a drawn dpf of 6 from Example 1 I is broken and formed into two identical homogeneous webs in the manner of as in Example 2 H, to obtain a nonwoven, identified as NW-10, is then cut into convenient dimensions for testing purposes, and conventional test results reported in Table I below as a control.

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TABLE 1

| 5  | Example | Material<br>Sample | Bale<br>From<br>Ex. | Webs      | Cross<br>Section<br>:0 | Length<br>(inches)<br>:0 | Opacity*4 in % | Feel* <sup>3A</sup>     | CD*5<br>Dry(gms) |
|----|---------|--------------------|---------------------|-----------|------------------------|--------------------------|----------------|-------------------------|------------------|
|    | 2 A     | NW-1               | 1A                  | Same      | 100:0                  | 1":0                     | 41             | Coarse                  | 382              |
|    | 2B*3    | NW-2               | 1B                  | Same      | 0:100                  | 0:2"                     | 26             | Excellent               | 424              |
|    | 2 A/B   | NW-3               | 1A                  | Different | 25:75                  | 1":2"                    | 32             | Excellent*7             | 447              |
| 10 |         |                    | 1B                  |           |                        |                          |                | Fairly Soft*6           |                  |
|    | 2 A/B   | NW-4               | 1A                  | Different | 50:50                  | 1":2"                    | 37             | Excellent*7             | 410              |
|    |         |                    | 1B                  |           |                        |                          |                |                         |                  |
|    | 2 E     | NW-5               | 1A                  | Different | 75:25                  | 1":2"                    | 39             | Fairly Soft*6           | 379              |
|    |         |                    | 1B                  |           |                        |                          |                |                         | ]                |
| 15 | 2 F     | NW-6               | 1B                  | Same      | 50:50                  | 2":2"                    | 35             | Soft                    | 454              |
|    |         |                    | 1C                  |           |                        |                          |                |                         | 1                |
|    | 2 G     | NW-7               | 1D                  | Different | 50:50                  | 1.5":1.5"                | 35             | Excellent*7             | 364              |
|    |         |                    | 1E                  |           |                        |                          |                | ·                       |                  |
|    | 2 H*3   | NW-8               | 1G                  | Same      | 0:100                  | 0:1.5"                   | 42             | Excellent               | 177              |
| 20 | 2 1 3   | NW-9               | 1H                  | Same      | 100:0                  | 1.5":1.5"                | 44             | Soft                    | 234              |
|    | 2 J*3   | NW-10              | 11                  | Same      | 0:100                  | 1.5":1.5"                | 23             | Coarse (like polyester) | 304              |

<sup>\*3</sup> Control.

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# Example 3

- A. Diamond cross-sectional isotactic polypropylene filament of 6.0 dpf spun denier is obtained in a conventional manner by melt spinning at 290°C. using PRO-FAXR 6501 polypropylene polymer, degraded, spun and processed in the manner of Example 1 A to obtain a final drawn denier of 2.1, then cut to 2.5 cm length, baled, and stored for later use.
- B. Delta cross-sectional isotactic polypropylene filament having a 2.6 dpf spun denier, is produced in the manner described in Example 1 C to a drawn denier of 2.1, then cut into 5 cm bundles and baled for later testing.
- C. Round cross-sectional isotactic polypropylene filament of 2.8 dpf spun denier is produced as described in Example 1 B to a drawn denier of 2.1 then cut into 5 cm bundles and baled for later testing.

#### Example 4

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Three test nonwoven samples are prepared as follows:

A. Nonwoven test strips are prepared by conventionally producing homogeneous webs varying in weight within a range of about 10-15 gm/yd², using filaments of diamond cross-section configuration from Example 3 A. Random combinations of two homogeneous webs, thus produced, are superimposed in machine direction onto a continuous fiber glass belt and bonded using a diamond-patterned calendar at 165°C and 276kPa (40 psi). The resulting nonwoven test materials are cut, weighed and tested for opacity using a Diano Match Scan II color spectrometer, and the results reported in Table II below as S-1, S-2 and

<sup>&</sup>quot;3A For evaluation purposes the term "Coarse" here denotes an unsatisfactory feel for commercial use as diaper coverstock and "Excellent" denotes a superior feel and softness acceptable for commercial usage, "Soft" denotes high qual ty commercially acceptable feel and softness while "Fairly Soft" denotes marginally acceptable feel and softness.

<sup>\*4</sup> An opaqueness of 39% or above is here considered commercially superior as diaper coverstock and 32% considered a modest though significant improvement.

<sup>\*5</sup> A CD dry strength of 300gm or higher is considered commercially acceptable as diaper coverstock.
\*6 Tested for softness on the delta cross-sectional side.

<sup>\*7</sup> Tested for softness on the circular cross-sectional side.

S-3.

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- B. Nonwoven test strips are prepared by producing homogenous webs varying in weight within a range of about 10-15 gm/yd² using the filaments of round cross-sectional configuration reported in Example 3 C. Random combinations of two homogeneous webs, thus produced, are superimposed in machine direction onto a continuous fiber glass belt and bonded using a diamond-patterned calendar at 165 °C. and 276kPa. The resulting nonwovens are cut, weighed and tested for opacity using a Diano Match Scan II Color Spectrometer, and the results reported in Table II below as S-10, S-11 and S-12.
- C. Nonwoven test strips are prepared by conventionally producing homogeneous webs varying in weight from about 10-15 gm/yd² using filaments of delta cross-sectional configuration reported in Example 3 B. Random combinations of two homogeneous webs thus produced are superimposed in machine direction onto a continuous fiber glass belt and bonded using a diamond-patterned calendar at 165 °C and 276kPa. The resulting nonwovens are cut, weighed and tested for opacity as before and test results reported in Table II as S-4, S-5 and S-6.
- D. Nonwoven test strips are prepared by producing homogenous webs of diamond and of delta cross-sectional configuration as in Examples 3 A and 3 B supra. Webs of different fiber cross section are randomly chosen, superimposed in machine direction, and bonded to obtain test nonwovens having 50%:50% by weight of diamond:delta-fiber content, then the nonwoven is cut, weighed and tested as before. Test results are reported in Table II below as S-7, S-8 and S-9.

TABLE II

| Sample | Fiber Content<br>Cross-Section<br>Configuration | Nonwoven wt<br>gm/yd² (2 webs) | Opacity % |
|--------|-------------------------------------------------|--------------------------------|-----------|
| S-1    | 100% Diamond                                    | 20.0                           | 37.0      |
| S-2    | 100% Diamond                                    | 21.5                           | 37.5      |
| S-3    | 100% Diamond                                    | 26.0                           | 40.5      |
| S-4    | 100% Delta                                      | 20.1                           | 41.0      |
| S-5    | 100% Delta                                      | 21.5                           | 42.2      |
| S-6    | 100% Delta                                      | 26.0                           | 46.0      |
| S-7    | 50% Diamond)<br>50% Delta )                     | 20.0                           | 40.5      |
| S-8    | 50% Diamond)<br>50% Delta )                     | 21.5                           | 41.0      |
| S-9    | 50% Diamond)<br>50% Delta )                     | 26.0                           | 44.0      |
| S-10   | 100% Round                                      | 20.2                           | 28.0      |
| S-11   | 100% Round                                      | 21.5                           | 29.5      |
| S-12   | 100% Round                                      | 26.2                           | 34.0      |

### Claims

- 1. A method for making nonwoven material containing polyolefin filaments, comprising assembling a web of filaments, including the polyolefin filaments, and bonding the filaments to form a nonwoven material, is characterized in that at least about 25% of the total weight of the filaments in the web are polyolefin filaments that have a delta "\( \Delta \)" or diamond cross-section configuration, an initial spun denier not exceeding about 24 dpf, and a final drawn denier of not less than about 1 dpf.
- 2. A method for making nonwoven material as claimed in claim 1, further characterized in that the polyolefin filaments have a initial spun denier within a range of about 24-6 dpf and a final drawn denier of not less than about 1.9 dpf.
- 3. A method for making nonwoven material as claimed in claim 1, further characterized in that the polyolefin filaments have an initial spun denier not exceeding about 4 dpf and a final drawn denier of not less than about 1 dpf.
  - 4. A method for making nonwoven material as claimed in claim 1, further characterized in that a

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polyolefin filament has an initial spun denier not exceeding about 6 dpf.

- 5. A method for making nonwoven material as claimed in claim 3, further characterized in that the polyolefin filament has an initial spun denier within the range of about 2.0-4.0 dpf and a final drawn denier above 1.9 dpf.
- 6. A method for making nonwoven material as claimed in claim 5, further characterized in that the polyolefin filaments have a final drawn denier above 2.5 dpf.
- 7. A method for making nonwoven material as claimed in any of the preceding claims, further characterized in that the nonwoven material comprises polyolefin filaments of mixed delta and round cross-sectional configuration.
- 8. A method for making nonwoven material as claimed in any of the preceding claims, further characterized in that the nonwoven material comprises polyolefin filaments of mixed delta and diamond cross-sectional configuration.
- 9. A method for making nonwoven material as claimed in any of the preceding claims, further characterized in that 50% of the nonwoven material comprises polyolefin filaments of mixed delta or diamond cross-sectional configuration and the remaining filaments have a round cross-sectional configuration.
- 10. A method for making nonwoven material as claimed in any of the preceding claims, further characterized in that the filaments used vary from about 2.5-7.6 cm in length.
- 11. A method for making nonwoven material as claimed in claim 10, further characterized in that the nonwoven material comprises 2.5 cm-long polyolefin filaments of diamond cross-sectional configuration and 3.8-5 cm-long round cross-sectional filaments.
- 12. A nonwoven material containing polyolefin filaments, whenever made by the method as claimed in any of the preceding claims, characterized in that at least about 25% of the total weight of the filaments in the web are polyolefin filaments that have a delta "\Delta" or diamond cross-sectional configuration, an initial spun denier not exceeding about 24 dpf, and a final drawn denier of not less than about 1 dpf.

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- Soft water-permeable polyolefins nonwovens having opaque characteristics.
- ⑤ A method for making nonwoven material containing polyolefin filaments and having an increased degree of opacity, characterized in that at least about 25% of the total weight of the filaments in the nonwoven web are polyolefin filaments that have a delta "Δ" or diamond cross-sectional configuration, an initial spun denier not exceeding about 24 dpf, and a final drawn denier of not less than about 1 dpf is disclosed.

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# EUROPEAN SEARCH REPORT

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| of relevant<br>EP-A-296572 (HERCULE                              |                                                                                                        |                                                                                                                                                                                                                                                                                             | CLASSIFICATION OF                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  |
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|                                                                  |                                                                                                        | to claim                                                                                                                                                                                                                                                                                    | APPLICATION (Int. CL                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |
| * column 1, line 41 -                                            | o inc.)<br>- column 2, line 51; claims                                                                 | 1, 3-6,<br>10, 12                                                                                                                                                                                                                                                                           | DO4H1/42                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           |
| 1-9 *                                                            | •                                                                                                      | 2 7 0                                                                                                                                                                                                                                                                                       |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |
|                                                                  |                                                                                                        | 2, 7-9,<br>11                                                                                                                                                                                                                                                                               |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |
| GB-A-1086873 (I.C.I.)                                            | <del></del>                                                                                            | 1, 70                                                                                                                                                                                                                                                                                       |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |
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|                                                                  | een drawn up for all claims                                                                            |                                                                                                                                                                                                                                                                                             |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |
| 1                                                                | . Date of completion of the search                                                                     |                                                                                                                                                                                                                                                                                             | Examiner                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           |
|                                                                  |                                                                                                        | 1                                                                                                                                                                                                                                                                                           |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |
|                                                                  |                                                                                                        | underlying the in-<br>ument, but publishe                                                                                                                                                                                                                                                   | vention<br>ed on, or                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |
| liarly relevant if combined with and<br>ent of the same category | other D: document cited in<br>L: document cited for                                                    | the application<br>ther reasons                                                                                                                                                                                                                                                             |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                    |
|                                                                  | * page 1, line 20 - p  * The present search report has believed search HE HAGUE TEGORY OF CITED DOCUME | TEGORY OF CITED DOCUMENTS  T: theory or principle F: earlier patent document cited in a first the filing date ent of the same category logical background  T: theory or principle F: earlier patent document cited in a first the filing date II: document cited in II.: document cited for | GB-A-1086873 (I.C.I.)  * page 1, line 20 - page 2, line 99; claims 1, 2  *  The present search report has been drawn up for all claims  The present search report has been drawn up for all claims  Place of search  IE HAGUE  Date of completion of the search  E HAGUE  TEGORY OF CITED DOCUMENTS  Place of the same category  Description of the same category  Description of the same patent document, but publish  Test theory or principle underlying the in  E cattler patent document, but publish  D : document cited in the application  D : document cited in the application  D : document cited in the application  A: member of the same patent family.  A: member of the same patent family.  A: member of the same patent family. |